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P.17

LARGE AREA NUCLEAR PARTICLE DETECTORS USING ET MATERIALS

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QF 572551

PROJECT SUMMARY:

Purpose: The purpose of this SBIR Phase I feasibility effort was to demonstrate the usefulness of Quantex electron-trapping (ET) materials for spatial detection of nuclear particles over large areas. This demonstration entailed evaluating the prompt visible scintillation as nuclear particles impinged on films of ET materials, and subsequently detecting the nuclear particle impingement information pattern stored in the ET material, by means of the visible-wavelength luminescence produced by near-infrared interrogation. Readily useful levels of scintillation and luminescence outputs were to be demonstrated.

Research Carried Out: The specific tasks performed were to:

1. Fabricate sample detector planes of ET materials.
2. Assemble measurement apparatus for providing alpha and beta excitation, IR interrogation and visible-wavelength readout of instantaneous and stored impingement information.
3. Measure the effects of alpha and beta particle impingement on ET detectors in providing trapped electrons by measuring the response to infrared probe pulses, including intensity, pulse duration, wavelength, reproducibility, position resolution and erasure.
4. Measure the prompt visible scintillation intensity under alpha and beta particle impingement.
5. Determine the radiation hardness of ET materials.
6. Evaluate the performance of ET materials vis-a-vis other large-area nuclear particle detectors.

Results: Useful levels of both scintillation and storage for later readout were successfully achieved for both alpha and beta particle impingement. Permanent damage effects were negligible at 100 kilorad accumulated dose for beta irradiation. The measured infrared-addressed storage is linear with accumulated particle impingement. Video readout is quite readily feasible with available image intensifiers and silicon camera chips.

Potential Applications: Large-area energetic particle detection with built-in storage for rapid readout upon command. Industrial or medical radiography with laser-diode readout of stored images. Biomedical radio tracer mapping in electrophoresis separation.

(NASA-CR-180777) LARGE AREA NUCLEAR PARTICLE DETECTORS USING ET MATERIALS (Quantex Corp.) 17 p CSCL 14B

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I. PROJECT OBJECTIVES:

In order to demonstrate the feasability of ET materials as imaging nuclear particle detectors, the specific Phase I objectives were to:

1. Demonstrate the photonic characteristics of ET materials, i.e., stimulated luminescence wavelength spectrum, IR stimulation wavelength sensitivity, etc.

2. Demonstrate the ability of energetic nuclear particles to create trapped electrons in large area ET detector configuartions which can then be released, sending proportionate visible photons to an optical detector.

3. Demonstrate the combined availability of instantaneous and time-integrated nuclear particle impingement locations.

4. Assess the usefulness of ET materials as large area nuclear particle detectors integrated with video readout.

5. Demonstrate the radiation hardness of ET detectors.

6. Recommend potential compounds and technologies for Phase II Research.

II. DESCRIPTION OF THE WORK PERFORMED:

The specific tasks performed to achieve the project objectives were as follows:

A. ET Detector Fabrication

The first step was to scan through various previously developed formulations of Quantex ET materials to find the most promising response to high-energy particle impingement. This pre-sorting process entailed contact exposure of bulk ingot surfaces to emissions from a 0.5 microCurie ^{241}Am isotope source and subsequent illumination with near-infrared to see the qualitative amount of electron trapping storage produced. By far the most sensitive were the Quantex-proprietary formulations of the group internally called Q-16.

A pre-surveyed ingot of this formulation was then reduced to powder form and seived to produce a 20 to 75 micrometer particle size distribution. The powder particles of this formulation were then settled out of a liquid suspension onto optical-quality sapphire substrates and treated at elevated temperatures (by a previously developed Quantex process) to directly bond the active ET particles to the substrate. Excellent adhesion was obtained. This technique was chosen for these general-purpose experimental samples to avoid interference from encapsulating binders or coatings for shallow interaction depths, e.g., as

with alpha particles. The resulting thickness of the ET films was approximately 250 microns.

B. Scintillation/Storage Measurement Apparatus

The measurement apparatus is shown schematically in Figure 1. The alpha and beta sources employed were 5.3 MeV ^{210}Po and 0.55 MeV ^{90}Sr nuclides respectively, both of about 0.1 microCurie activity level. These sources were small volumes of the isotopes bonded into shallow cavities in 1-inch diameter substrates, and were obtained from The Nucleus Co.,of Oak Ridge Tennessee, after surveying various available supplies (such as at Amersham Corporation, etc.) The ET samples were placed directly on the photomultiplier housing for good light capture . The housing was configured to contain a 650 nm wavelength short-pass filter to block near-infrared, and a Hamamatsu R268 photomultiplier tube. The isotope sources were placed directly against the ET samples to minimize air interaction. An infrared emitting diode (IRED) of 960 nm wavelength was used to provide the stimulation for releasing trapped electrons to produce visible luminescence. The diode output was controlled by setting its current with a regulated constant-current power supply. The IR intensity delivered by the diode at specific drive currents was measured with a calibrated silicon photodiode of linear photocurrent vs. intensity, obtained from EG&G, and a precision Kiethley picoammeter. As a measure to keep extraneous light and IR from reaching the ET sample and the photomultiplier tube, the entire sensing apparatus was enclosed in a box.

The response of the photomultiplier tube was calibrated against the EG&G photodiode, by using calibrated neutral-density filters to allow attenuation of known input light intensities to the levels of the scintillations and stimulated emissions being measured. The PMT output current pulses during the scintillation or the stimulated luminescence were captured with a storage oscilloscope.

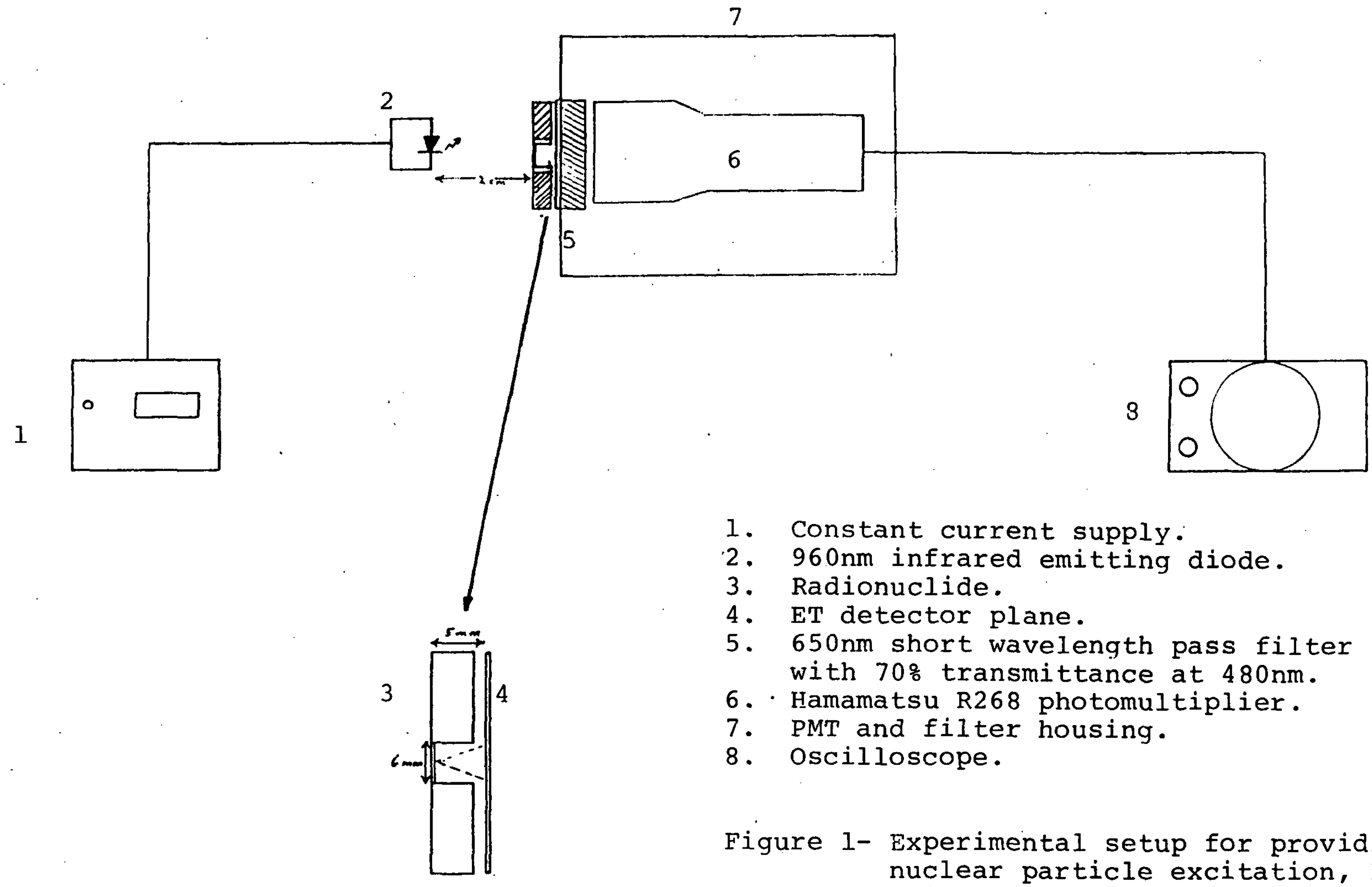
C. Measurements

1. Stimulation Spectrum

The trapped electron stimulation spectrum was measured to determine the optimum IR wavelengths to use for stimulating visible emission from the ET detector planes. This was done by first generating trapped electron populations in the samples with fixed exposures to alpha particles and then addressing them with constant intensities at different IR wavelengths. A plot was then made of relative visible emission intensity versus IR wavelength.

2. Luminescence Spectrum

The visible luminescence spectra of the ET samples were measured with a Licor-1800 spectroradiometer during infrared illumination. Knowing the peak luminescence wavelength and bandwidth was required for choosing an appropriate photomultiplier.



1. Constant current supply.
2. 960nm infrared emitting diode.
3. Radionuclide.
4. ET detector plane.
5. 650nm short wavelength pass filter with 70% transmittance at 480nm.
6. Hamamatsu R268 photomultiplier.
7. PMT and filter housing.
8. Oscilloscope.

Figure 1- Experimental setup for providing nuclear particle excitation, IR stimulation, luminescence detection and measurement of ET detector planes.

3. ET Detector Response

a. Scintillation

The prompt scintillation resulting from nuclear particle impingement on the ET detector planes was measured by placing the alpha and beta sources on the ET sample (see Figure 1) and measuring the PMT response to the emissions coming through the transparent substrate. In this way, both the frequency of scintillation events and the magnitude of each event response was determined.

b. Electron Trapping

The efficacy of nuclear particle impingements creating trapped electron populations in the ET samples was determined by exposing them to incremented doses of incident particles and measuring the subsequent IR stimulated luminesence (IRSL) intensity with the photomultier. The visible IRSL emission was measured as a function of time under continuous IR stimulation until exhaustion, to determine the total storage for each nuclear particle dose. Also, the IRSL output intensity was measured as a function of IR input intensity.

In addition, a measurement of the background signal level was made before each nuclear particle exposure. Variations in the background appeared to arise from residual trapped electron populations in the ET film not fully erased between successive measurements and PMT response to residual IR not completely blocked around the edges of the the short-pass filter. These very small background signals were subtracted from the data obtained so as not to impose these artifacts on the plotted results.

c. Trapped Electron Population Retention

Measurements were also carried out to assess any changes in the trapped electron population over extended time periods. This was done by exposing ET detector planes to doses of nuclear particles and measuring the IRSL at progressively longer times after the exposure.

d. Particle Impingement Image

In addition to quantitative photonic measurements, the imaging capabilities of ET materials was assessed. This was done by first exposing our ET detector planes to the radiation being emitting by the isotope sources. Next, the ET detector plane was placed in contact with photographic film and exposed to IR projected through the sapphire substrate. Thereby the IRSL emission pattern was recorded on the film, showing the nuclear particle impingement pattern. (A special film not sensitive to IR wavelengths was used so that the recorded patterns were due to the visible luminesence from the ET material, and not the IR excitation.)

4. Radiation Hardness

The stability of ET detectors in radiation environments was assessed by evaluating the integral of the IRSL output before and after exposure to a 0.1 Mrad dose of 350 KeV electrons. The available integral was evaluated by first filling traps with a known exposure to short wavelength (450nm) light. The integrated luminescence was then determined by continuous exposure to IR. Any changes in active storage trap density due to possible radiation damage would be manifested in a change in the integrated luminescence after irradiation.

The high-dose electron irradiation and associated dosimetry were performed at the National Bureau of Standards in Gaithersburg, Maryland, by the resident NBS staff as a subcontracted activity.

D. Comparison to Other Large Area Detectors

One of the tasks in the contract effort was to compare the capabilities of ET detector planes to other large-area detectors. Increasing the area of a detector increases the probability that low spatial frequency events will be detected. Also, if the detector medium has the capability for storing event information, the position of events can be determined without a requirement for precise coincident determinations.

A very wide range of detectors has been developed for a wide variety of terrestial and space-related applications. For space-related applications there are some significant constraints, among which is the use of solid-state materials because interaction volumes are minimized and overall system complexity is reduced. Consequently, the candidate detectors are either 1) semiconductor detectors in which the radiation absorption and detection occur in the same material (e.g., silicon or germanium diodes) or 2) scintillator media mated with photomultiplier tubes, in which configuration light is produced in the scintillator and is detected by the photomultiplier. Nuclear emulsions and radiographic films, though large area detectors, are not considered because the stored information would not be easily retrieved.

Semiconductor detectors generally have been the detectors of choice for particulate radiation, because the range of the particles is usually less than the depletion region depth in the detectors. They have good energy resolution, excellent timing characteristics, good stability and simplicity of operation. However, detector area appears to be limited to $20cm^2$ or less. Larger areas, therefore, can only be achieved if individual detectors are arrayed. However, arraying such detectors is relatively costly. For this reason, the use of large area scintillators mated to photomultiplier tubes is likely to be the most economic alternative.

Plastic scintillators, in particular, can be fabricated in any shape or size, have low absorption of their output luminescence, are fast, and are relatively inexpensive. Some disadvantages are the relatively low scintillation efficiencies, poorer energy resolution, low density and low effective atomic number. It is clear that it would be advantageous to have a scintillator that had a higher efficiency, higher density and higher effective atomic number while maintaining the capabilities of being fabricable in large areas, having low self absorption, being fast and inexpensive. Further, the ability to store information in the scintillating media for later retrieval would have definite advantages for certain applications.

III. RESULTS:

1. Trapped Electron Stimulation Spectrum

Figure 2 shows the IR sensitivity spectrum for stimulating trapped electrons to luminescence for the ET samples employed in this effort. On this curve the optimum IR wavelength for stimulating trapped electrons occurs at 980 nm. The IRED chosen for these experiments had its peak emission at a wavelength of 960 nm, the response to which is within 10% of the peak in Figure 2, and was quite suitable for stimulating visible luminescence in the samples.

2. Luminescence Spectrum

Figure 3 shows the IRSL output spectrum as measured with a Licor model 1800 spectroradiometer. The peak luminescence wavelength occurs at approximately 480 nm.

A Hamamatsu R268 photomultiplier tube exhibits its greatest spectral sensitivity in the wavelength range shown in Figure 3, and was found to be suitable for the various measurements of IRSL output. Also, this luminescence spectrum is well suited for image recording using photographic film or optoelectronic imaging equipment.

3. ET Detector Response

a. Scintillation

Since the ET samples were fabricated with relatively arbitrary thickness and we used a substrate-side emission measurement system, two additional steps were taken to check scintillation measurements:

1) A thin P-22 (commercially available phosphor) screen was fabricated and also used for checking scintillation counting with the same measurement apparatus. This screen consisted of a 10 mg/cm^2 layer of P-22 phosphor on a suitable transparent substrate and was aluminum coated (to a non-interfering 300 Angstrom thickness) to enhance light

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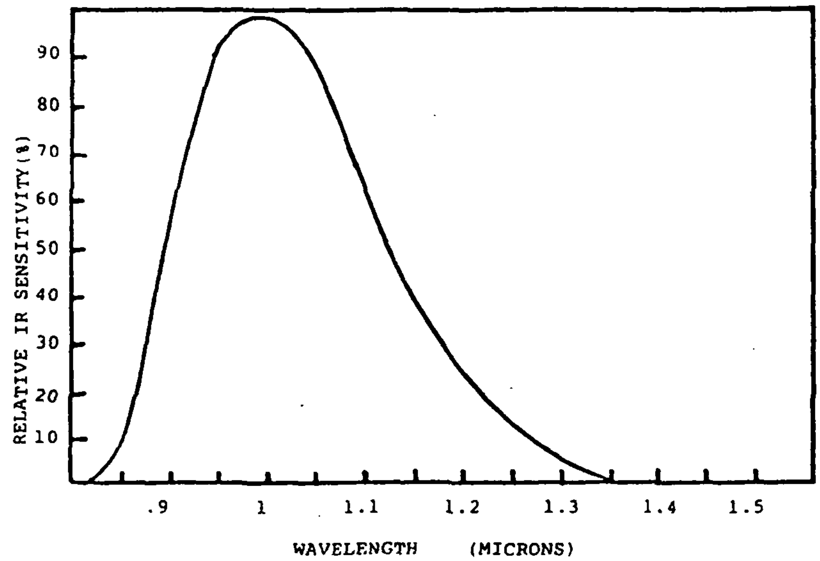

Figure 2- IR stimulation wavelength sensitivity.

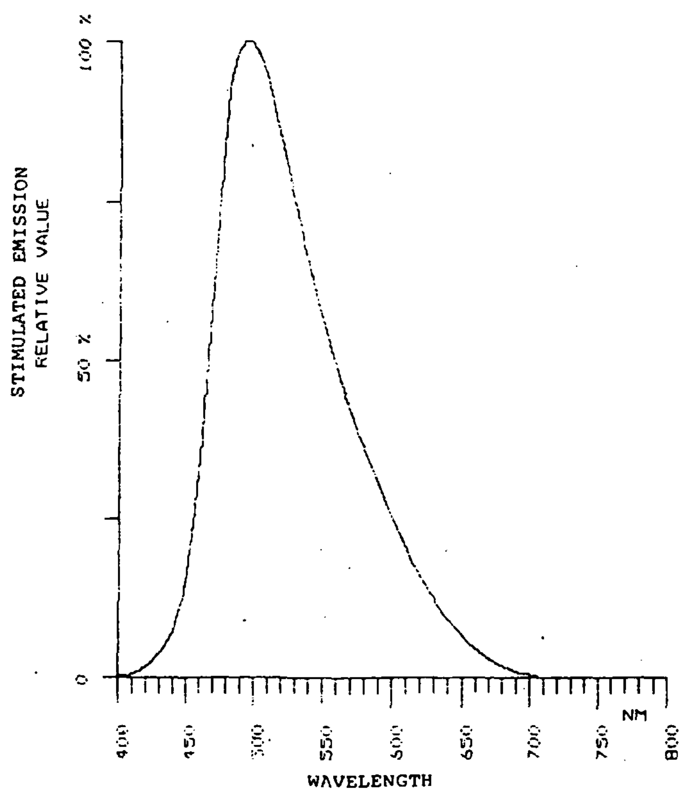

Figure 3- IR stimulated luminescence wavelength spectrum.

coupling to the PMT. The P-22 screen produced light pulses far above the noise of the measurement system, at rates of 800 counts/second for the ^{210}Po alpha source and 600 counts/second for the ^{90}Sr beta source. The back-side light pulse intensity was a factor of 10 greater for the thin P-22 phosphor screen than for the thicker ET samples.

2) The light transmission at the emission wavelengths was measured from the front surface of the ET samples through to the back of their substrates. The transmission of the samples at these wavelengths was found to be almost exactly 20%. Therefore, it was apparent that 80% of the IRSL light generated in the shallow alpha-absorption-depth layer at the front side of the samples was not seen by the photomultiplier at the back of these samples. The actual IRSL output at the front surface, (or as would be seen at the back of a thin layer), was five times as great as measured on the photomultiplier side.

The scintillation pulses from particle impingement on the ET detector planes were not as far above the measurement system noise level, and were at rates of 400/second and 300/second, respectively, as counted directly from the oscilloscope traces for the sub-microCurie alpha and beta sources employed in the measurements. It is apparent that some half of the light scintillation pulses fell below the measurement threshold, as compared to the thin P-22 scintillator.

These results confirmed that the somewhat arbitrary thickness of the ET detector, which was 10-20 times greater than the P-22 layer, was far from optimum for scintillation counting of alpha and beta particles in the backside measurement arrangement. (Also, these initial-effort ET samples did not have the aluminum coating which improved the P-22 optical coupling.) A Phase II continuation effort would surely optimize the ET material thickness, reflective overcoats could be added, etc.. The important result here is that sufficient scintillation is apparently <u>generated</u> despite the energy input to storage, and further configuration improvements could indeed be made under Phase II efforts.

b. Electron Trapping

Figure 4 shows the IRSL output intensity versus the number of nuclear particle impingements obtained by the method described in section III above. The number of nuclear particles incident on the ET detector planes was determined by the scintillation rate of the P-22 phosphor screen.

Figure 4 shows a good correlation between the number of incident alpha and beta particles and the IRSL intensity. Also, it is seen that the ET storage responds in a linear fashion to both alpha particles and beta particles, a very important result.

Figure 5 shows the change in IRSL intensity under a continuous IR exposure of 100 microWatts/cm^2. The curves were obtained after exposure to 2×10^6 beta particles and 1.2×10^6 alpha particles. From these curves, the integrated luminescence output may be determined by calculating the area under each curve. This may be done in a straight forward manner

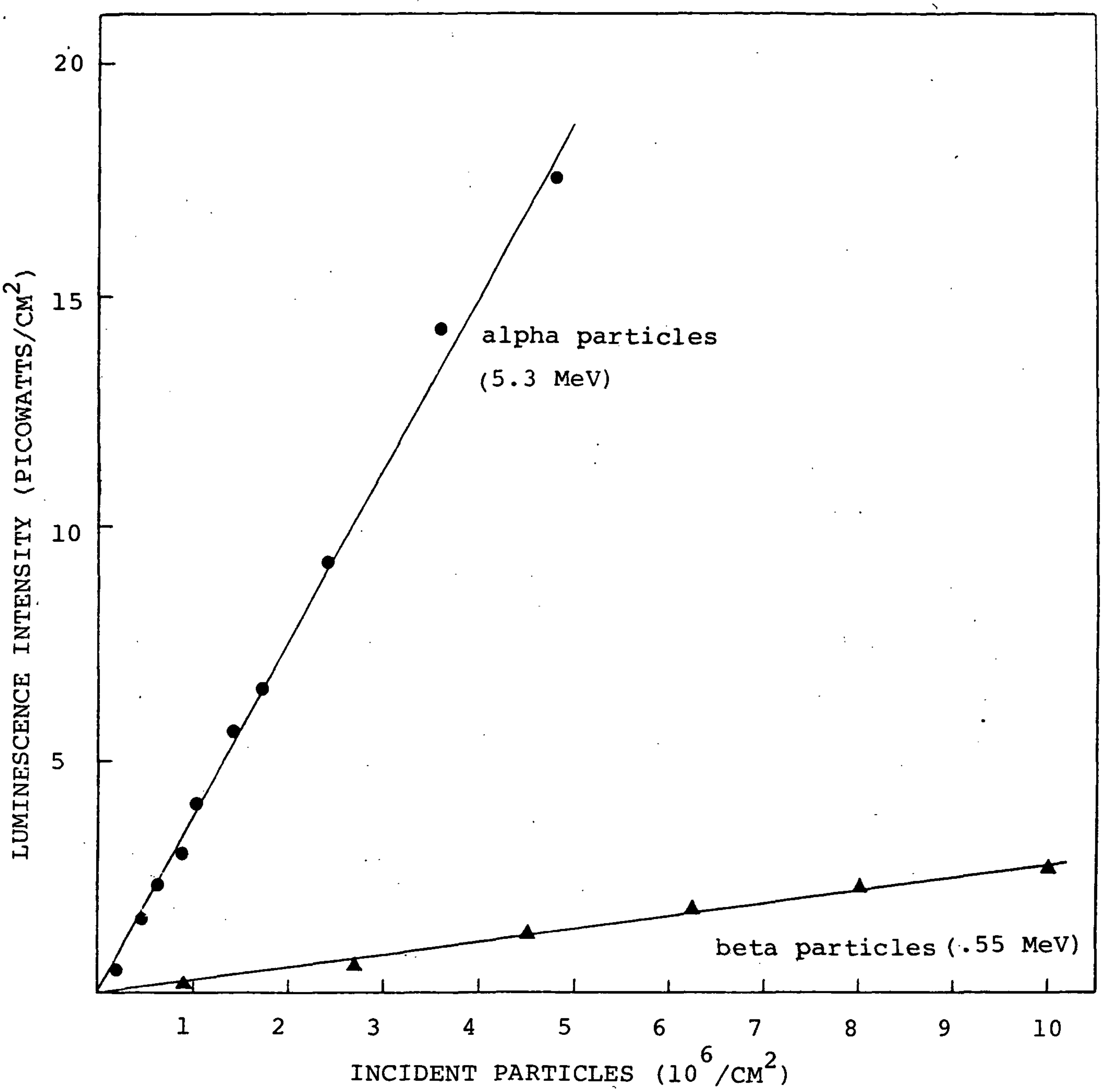

Figure 4- IR stimulated luminescence intensity following exposure to alpha and beta particles; IR intensity was 100 microwatts/cm^2.

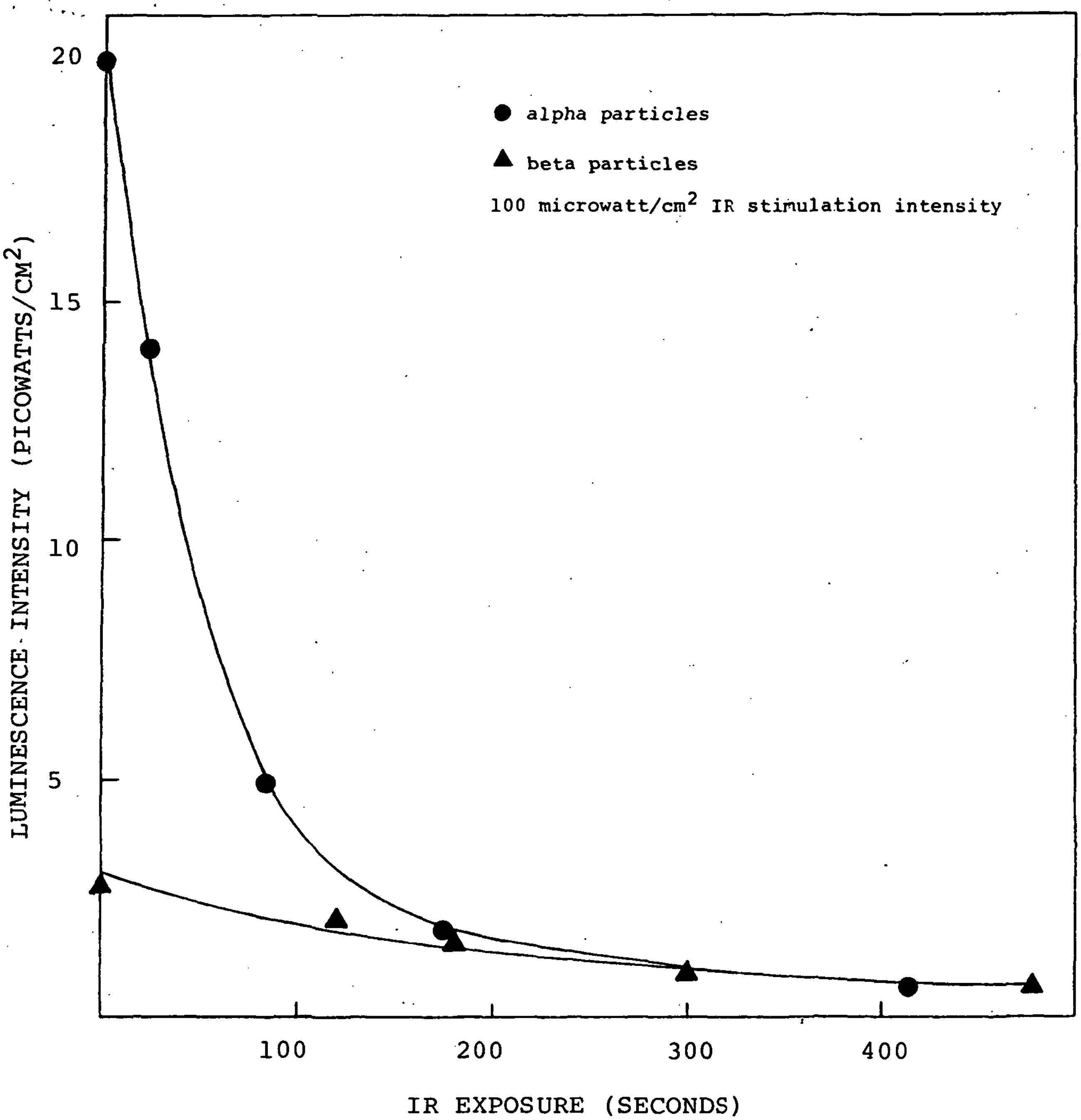

Figure 5- Decay of luminescence intensity under continuous stimulation by IR following exposure to $2X10^6$ beta particles and $1.2X10^6$ alpha particles.

by assuming the IRSL decreases according to $L = L_o \exp - t/\tau$, where L is the IRSL intensity at a time t, L_o is the initial IRSL intensity and τ is the IRSL intensity decay time to 1/e of its initial value. The integrated luminescence output is then $E=0.63L_o\tau$. Using appropriate values from figure 5, the integrated luminescence output following exposure to $1.2x10^6$ alpha particles was $2x10^{-10}$ Joule. Since the energy of photons of 480 nm wavelength is $4x10^{-19}$ Joule, 400 photons/alpha particle were measured by the photomultiplier tube upon IR stimulation. Correcting for the measured film transmission (5X), the IR-blocking filter's 70% transmission to the photomultiplier tube (1.4X) and the oppositely directed half of the output at the front surface (2X), the number of IR-generated photons per alpha particle is seen to be approximately 5600. A similar analysis for the beta particle data of Figure 5 results in back-surface response of 100 photons per beta particle, and a generated quantity of approximately 1400. Obviously, developing a thin film with a reflective overcoat would be of significant advantage.

Another important property of ET detector planes is that the maximum IRSL output, L_o, increases linearly with IR input intensity as shown in Figure 6. This means that the IRSL decay time, τ , decreases as the IR input intensity is increased. Therefore, this effect may be used to determine the IR intensity required to produce a sufficiently large L_o, yet a long enough τ , to allow for unambiguous signal detection in various optoelectronic systems.

c. Retention

Figure 7 shows a measured IRSL intensity resulting from exposure to $9.6x10^5$ alpha particles, sampled over extended periods. This curve indicates that after 10 hours approximately 50% of the original level is obtained, and that after 64 hours the sampled output decreased to 25% of its original value. It should be mentioned that Figure 7 represents data from very shallow alpha particle interaction with a powder sample, whereas other Quantex ET samples have shown much longer retention of trapped electrons stored by other inputs. It is fully expected that serious experimentation under a Phase II effort with smooth thin-films of ET material and surface-passivating overcoats to reduce surface recombination would yield greatly enhanced retention.

d. Particle Impingement Image

The quantitative data presented in previous sections shows that sufficient luminescence output is produced from ET materials to allow for image recording. Unfortunately, the limited resources available under this Phase I feasability study did not allow us to obtain the rather high-cost image intensifier apparatus to provide sufficient input to commercial-level video imaging equipment compatible with the 2800 emitted visible photons per alpha particle impact site that could be liberated by broad-area IR irradiation of the front surface. Common video camera chips require a minimum of 15,000 photons per pixel element, which could easily be supplied by a 10^6 gain microchannel-plate image intensifier intercepting a moderate solid angle of the emission. Such imaging could readily be pursued in a Phase II effort.

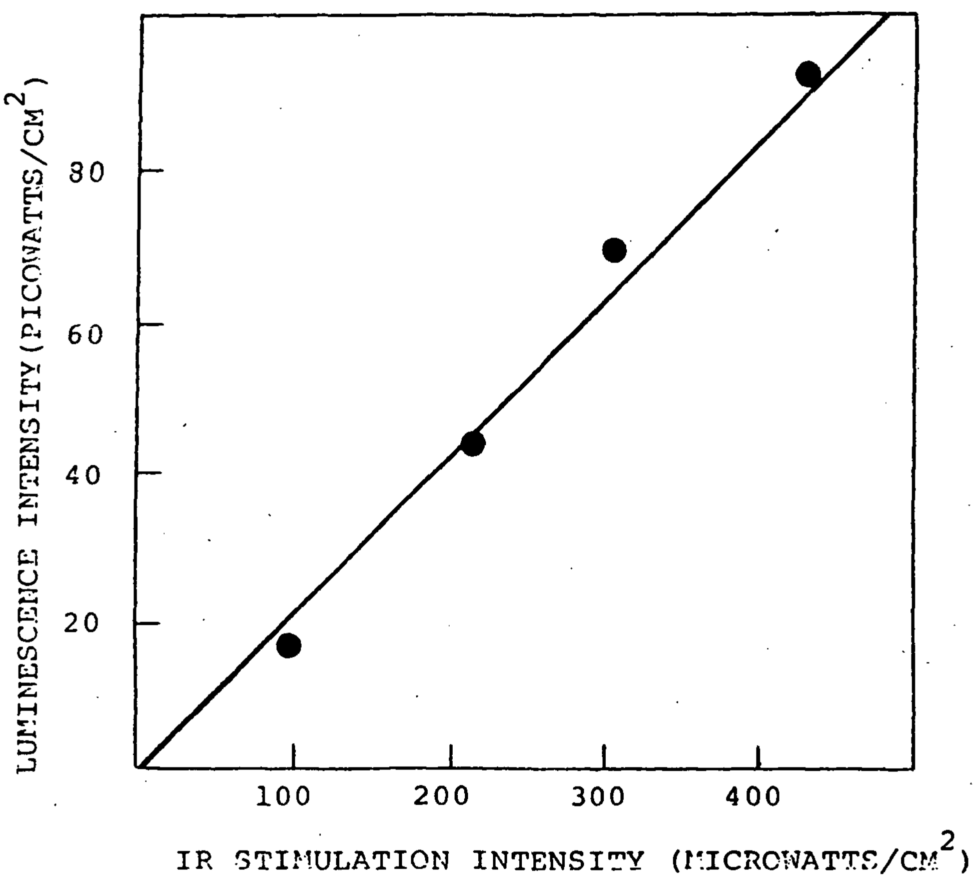

Figure 6- IR stimulated luminescence intensity versus IR stimulation intensity following exposure to 9.6×10^5 alpha particles.

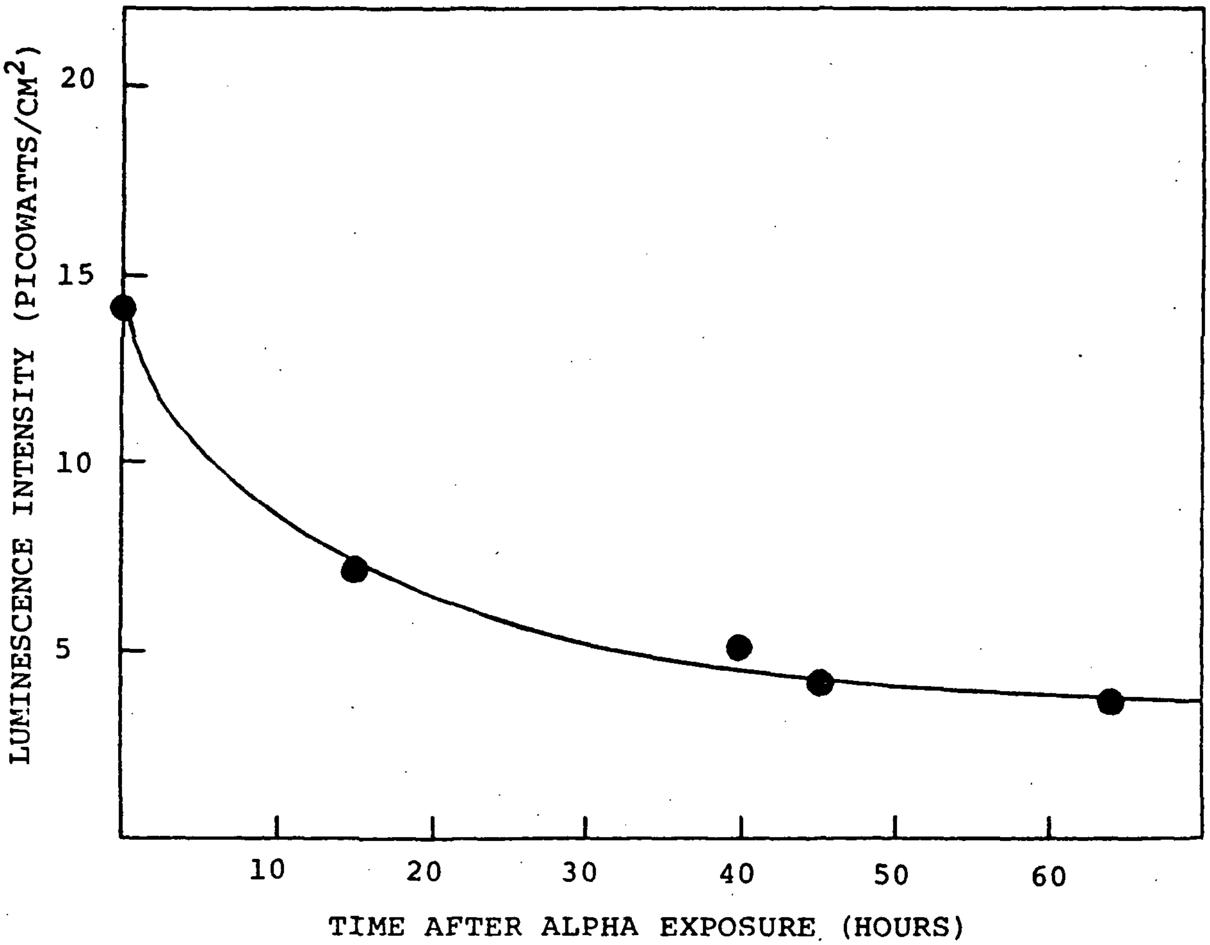

Figure 7- Retention of luminescence intensity over extended time periods; measurements were made following exposure to 9.6 X 10^5 alpha particles using 100 microwatt/cm^2 intensity IR.

However, images were recorded as contact prints on Polaroid Type 57 film. Several such images are shown in Figure 8. The pattern shown is the actual image of the ^{210}Po radionuclide source's emission pattern stored on ET detector planes and subsequently transferred as visible-light IRSL patterns upon IR illumination. Although it is difficult to quantitatively assess the resolution, it is readily possible to see the inhomogeneities present in the ^{210}Po source which were less than 1mm in size.

4. Radiation Hardness

The results of integrated luminescence measurements for identical storage before and after 0.1 Mrad of 350 KeV electron irradiation showed less than a 5% change, which is within the day-to-day set-up-error of the overall measurement system. (Of course a good deal of erasure had to be done before the second measurement cycle could commence.) This is consistent with previous studies on slightly different Quantex ET material samples, which also showed little or no measurable changes in the trapping capability, even following irradiations up to 1 Mrad.

IV. CONCLUSIONS AND PHASE II RECOMMENDATIONS:

The results of this Phase I effort have definitely demonstrated Technical Feasibility. The results obtained clearly indicate that ET materials can indeed be used for obtaining quantitative information about nuclear particle events with respect to both their quantity and spatial distribution. Moreover, it was shown that per alpha particle the ET material was capable of producing a sufficient number of trapped electrons to result in at least 400 photons actually being measured upon IR stimulation, and a total number of photons generated more like 5600 per particle. The respective photon quantities for beta particles were 100 and 1400.

The measurement system was purposely kept simple for this Phase I feasibility study, since a front-surface optical system was not applicable to either the mount configuration of the readily available alpha and beta isotope sources or atmosphere in the particle path. Due to visible light attenuation through the specific samples (80%) and the available IR-blocking filter (30%), and the isotropic emission of the ET material, a large majority of the IR-stimulated photons were not collected by the PMT behind the samples. Although the optical losses to back-side readout can not be completely eliminated, application of appropriate anti-reflection and reflection coatings to samples of optimized thickness would certainly result in greatly improved optical coupling.

Front-surface emission of 2800 photons per alpha particle represents a signal that will be detected with any reasonable PMT or image intensifier system. What will determine whether these photons can be easily detected is their rate of emission, which is in turn set by the incident IR stimulating intensity, and the solid intercept angle of an imaging system.

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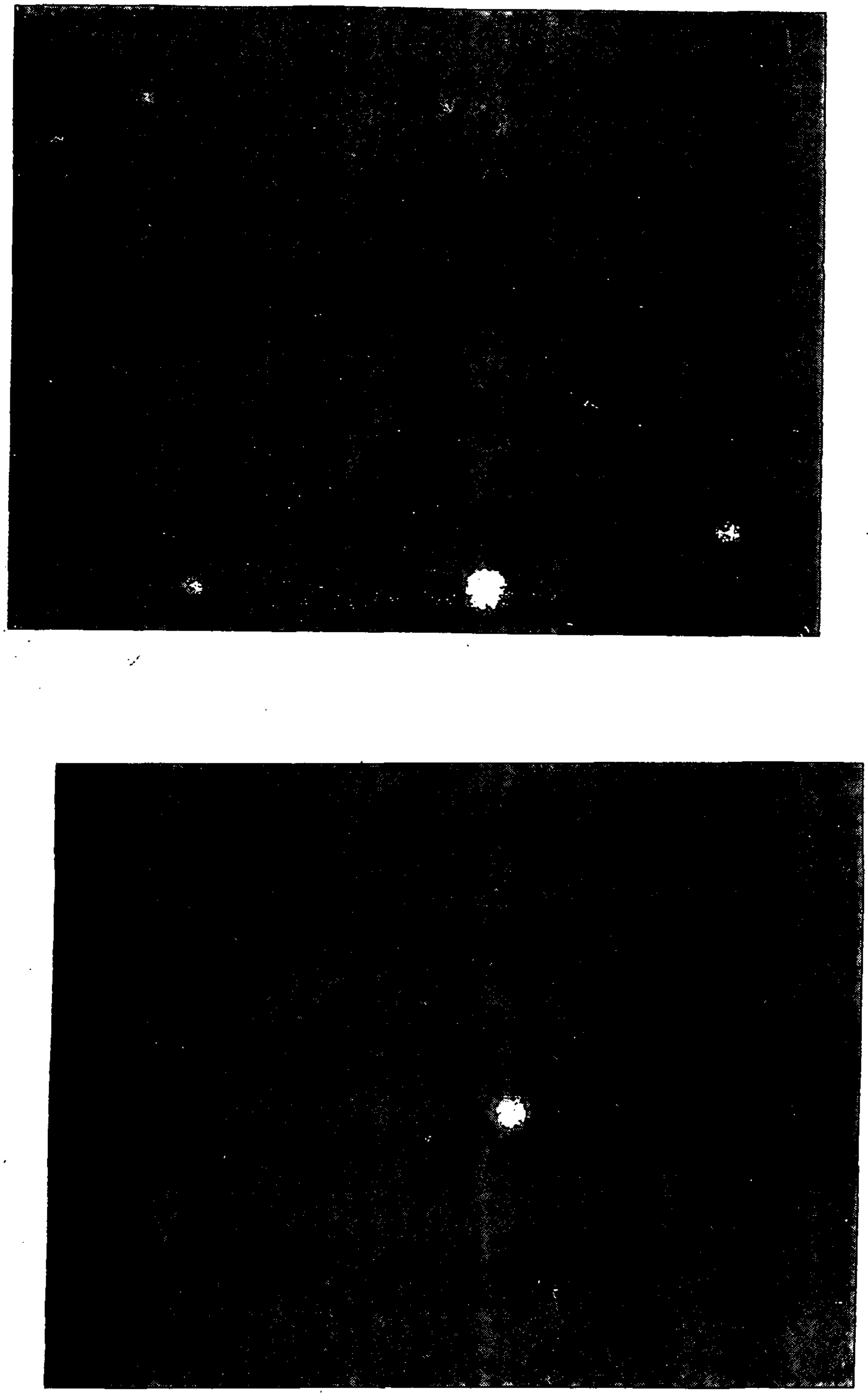

Figure 8 - The photographs above show alpha emission pattern produced by the ^{210}Po alpha source. These were stored on the ET detector plane and contact printed onto Polaroid Type 57 film during IR-stimulated visible luminescence. (Full size).

The IR intensity required to produce an L_o greater than the system detectability and also a τ longer than the system response time, may be calculated using the data shown in Figures 5 and 6 and by assuming $E=0.63L_o\tau$. The detectability of the photomultiplier system used in this effort was approximately 170 femtoWatts (with an "erased" ET sample in place) and the system response time was around 100 nanoseconds. Commonly available microchannel-plate image intensifiers easily surpass these levels, and also have higher available electronic gain.

Therefore, comfortably picking τ=100 microseconds and using the energy of 2800 emitted photons (1.1×10^{-15} Joule) one calculates that L_o is 11 picoWatts; a readily measurable level. In order to determine the IR intensity required to produce this output, the data in figure 5 was extrapolated to find L_o for 1 incident alpha particle at the specific IR intensity of 100 microwatts/cm^2. This value is then ratioed using Figure 6 to result in an IR intensity requirement of approximately 50 W/cm^2. A 50 Watt/cm^2 IR intensity is easily obtained from either commercial YAG lasers or junction-diode lasers. This intensity will not produce significant heating or other untoward effects in 100 microseconds. Interception of a relatively large fraction of the emitted photons would only require reasonable lenses to focus onto an intensifier photocathode. The gain available in common intensifiers would then supply image intensities far in excess of the minimum required for good contrast.

It is apparent that development of ET detector planes for practical application should take the following path in a Phase II program:

1. Configurations should be optimized for particle range-energy relationships, e.g., the thickness of the ET film should be much reduced as compared to that of the samples employed in Phase I. (Quantex is already starting thin film development for other applications of ET materials.)

2. Optical and surface-passivation coatings should be developed and applied to improve optical coupling and surface recombination.

3. Electronic imaging with image intensifiers and video camera chips, or spot scanning with electro-optic deflection, should be exercised with appropriate optics.

4. Quantification of response to other inputs should be explored, such as storing patterns of ultraviolet Cerenkov radiation from relativistic particle interactions.